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Material Attractiveness and Why It Is Important

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The Ohio State University
February 19, 2014

Outline

- Background
- Goals and Objectives
- Description and Discussion of Figure of Merit (FOM)
- Attractiveness Themes:
 - Reactor-grade plutonium,
 - Denaturing,
 - Dilution,
 - ^{233}U & thorium fuel cycles.
- Conclusions

Nuclear's “F” Words

- **Fissile** – A nuclide that is capable of undergoing fission by interaction with slow neutrons. Alternatively, a fissile nuclide has a 0.0253-eV fission cross section > 100 b and a half life > 1 year. Then, ^{233}U , ^{235}U , ^{236}Np , ^{239}Pu , ^{241}Pu , $^{242\text{m}}\text{Am}$, ^{243}Cm , ^{245}Cm , and ^{247}Cm are fissile.^{a,b}
- **Fissionable** – A nuclide that is capable of undergoing fission for neutron energies < 10 MeV, but is not fissile.^b An example is ^{238}U , which fissions at neutron energies > 1 MeV.
- **Fertile** – A nuclide that is not fissile, but may be used to produce fissile nuclides.^b Examples are ^{238}U and ^{232}Th .
- **Fissible** – A nuclide that has a bare critical mass.^c
- **Fizzle** – Refers to a degraded yield relative to the design yield, generally thought of as resulting from preinitiation.

^a Private communication with Eric Pitcher, LANL, (January 4, 2012).

^b J.R. Lamarsh, *Introduction to Nuclear Reactor Theory*, Addison-Wesley Publishing, Reading MA (1966).

^c R.E. Kelly and E. D. Clayton, “Fissible: A Proposed New Term in Nuclear Engineering,” *Nuc. Sci. Eng.*, **91**, 481 (1985).

Other Important Terms and Concepts

- **Multiplication Factor** – The ratio of the number of fissions in a generation to the number of fissions in the immediately preceding generation, aka k_{eff} .
 - $k_{\text{eff}} < 1$ – The system is subcritical,
 - $k_{\text{eff}} = 1$ – The system is critical,
 - $k_{\text{eff}} > 1$ – The system is supercritical.
- **Bare Critical Mass** – The mass of a material in air at which $k_{\text{eff}} = 1$.
- **Material Attractiveness** – The relative utility of nuclear material for an adversary in assembling a nuclear explosive device, taking into account the time and potential difficulties with separation and/or conversion, if needed, to a usable form.

Other Important Terms and Concepts (cont'd)

- **Proliferation resistance** is that characteristic of a nuclear energy system (NES) that impedes the diversion or undeclared production of nuclear material or misuse of technology by the *Host State* seeking to acquire nuclear weapons or other nuclear explosive devices.
- **Physical protection (robustness)** is that characteristic of an NES that impedes the theft of materials suitable for nuclear explosives or radiation dispersal devices and the sabotage of facilities and transportation by *subnational entities and other non-Host State adversaries*.

Why is Materials Attractiveness Important to You and the USA?

- Terrorists pose a threat to the USA:
 - The US is a favored terrorists' target
 - Terrorists have declared a desire to obtain a nuclear weapons capability.
- Approximately ¼ of the OSU students will get jobs at nuclear power plants:
 - All nuclear power plants breed plutonium
 - Even reactor-grade plutonium is weapons usable, but requires processing.
- Approximately ¼ of the OSU students will get jobs at national laboratories:
 - National laboratories associated with the weapons complex or nuclear energy generate, store, or work with plutonium
- Approximately ¼ of the OSU students will get jobs at nuclear vendors (e.g., Westinghouse, GE, B&W, etc.):
 - Vendors design reactors that:
 - Generate plutonium
 - Store plutonium

The Attractiveness of Materials in Advanced Nuclear Fuel Cycles for Various Proliferation and Theft Scenarios[†]

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October 9, 2011

[†]*Nuc. Tech.*, **179**, 5 (July, 2012)

Goals and Objectives

- Reduce materials attractiveness in nuclear fuel cycles – both present and future.
- Communicate relative attractiveness of weapons-usable nuclear materials without revealing sensitive information
- Strengthen international nuclear safeguard and security
 - Reinforce concept of “graded approaches”
 - Prevent reductions in existing safeguards & security requirements
 - Correct false or misleading statements
 - Propose guidance for future nuclear fuel cycles

Primary factors of material attractiveness

- **Bare Critical Mass (Utilization)**
 - Affects the device size and transportability
- **Internal Heat Generation (Utilization)**
 - Affects device shelf life and stability
- **Intrinsic Neutron Rate (Utilization)**
 - Causes pre-initiation and yield degradation
- **Radiation Dose Rate (Acquisition)**
 - Affects adversary's ability to acquire raw nuclear materials and assemble device
- **Net Weight (Acquisition)**
 - Affects adversary's ability to acquire raw nuclear materials
- **Processing Time and Complexity (Processing)**
 - Affects adversary's ability to convert the nuclear material into a pure metallic form

The following Figure of Merit (FOM) applies to a wide range of potential adversaries.

- FOM₁ applies to a technically advanced proliferant nation (in purified metal form) or a dedicated sub-national group (in unpurified metal form):

$$FOM_1 = 1 - \log_{10} \left(\underbrace{\frac{M}{800}}_{\text{Size Factor}} + \underbrace{\frac{Mh}{4500}}_{\text{Stability Factor}} + \underbrace{\frac{N}{10} \left[\frac{D}{500} \right]^{\frac{1}{\log_{10} 2}}}_{\text{Acquisition Factor}} \right)$$

- M – bare critical mass in purified or unpurified **metal** form (kg)
 - h – heat content in purified or unpurified **metal** form (W/kg)
 - D – dose rate of specified object of mass N or an unspecified object of mass 0.2•M @ 1 m (rad/h)
- If using the FOM to conduct safeguards and physical protection analyses, FOM₁ should be used because it bounds the range of nuclear materials that can potentially be processed and fabricated into a nuclear explosive device by an adversary.

Where does the FOM fit into proliferation resistance and physical protection?

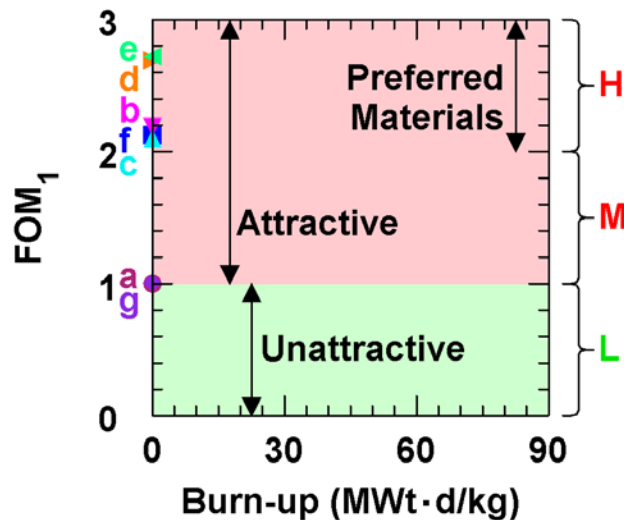
- Proliferation resistance measures, (host state):^a
 - Proliferation technical difficulty
 - Proliferation cost
 - Fissile material quality $\sim \text{FOM}_1$ (pure element only)
 - Proliferation time
 - Detection probability
 - Detection resources required
- Physical protection (sub-national state):^b
 - Material attractiveness $\sim \text{FOM}_1$ (impure alloy or pure element)
 - SNM quantity
 - Security category

^a Evaluation Methodology for Proliferation Resistance and Physical Protection of Generation IV Nuclear Energy Systems, GIF/PRPPWG-2006/005, <http://www.gen-4.org/Technology/horizontal/PRPPPEM.pdf>.

^b Nuclear Material Control and Accountability DOE M 470.4-6, <http://hss.energy.gov/nmmss/pdfs/m4704-6c1.pdf>.

The format of the FOM plots is given below.

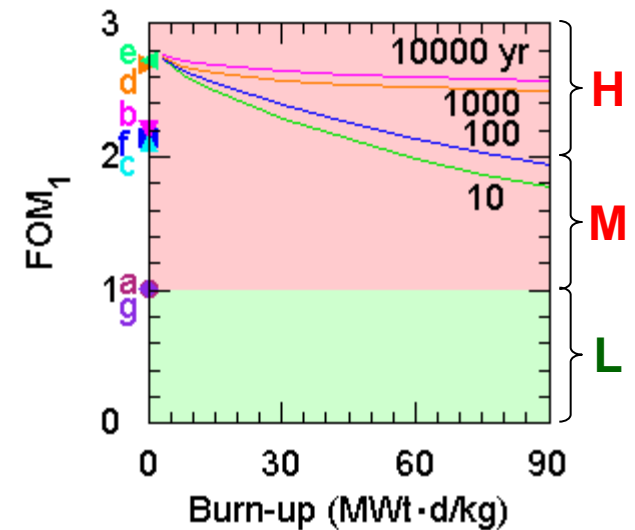
- The meaning of FOM_1 values:
 - $FOM_1 > 2$ (Red area): material is preferable for use in nuclear explosive devices
 - $FOM_1 > 1$ (Red area): material is attractive and should be safeguarded and secured
 - $FOM_1 > 0$ (Green area): material is unattractive, but may still be weapons usable
- The FOM_1 values of seven common materials (delineated in the blue box below) are shown for reference along the ordinate.



a – LEU (20% ^{235}U)
 b – HEU (93% ^{235}U)
 c – ^{237}Np
 d – ^{233}U (10 ppm ^{232}U)
 e – WG-Pu (94% ^{239}Pu)
 f – RG-Pu
 g – $^{238}\text{Pu}/^{239}\text{Pu}$ (80:20)

Reactor-grade plutonium is attractive.

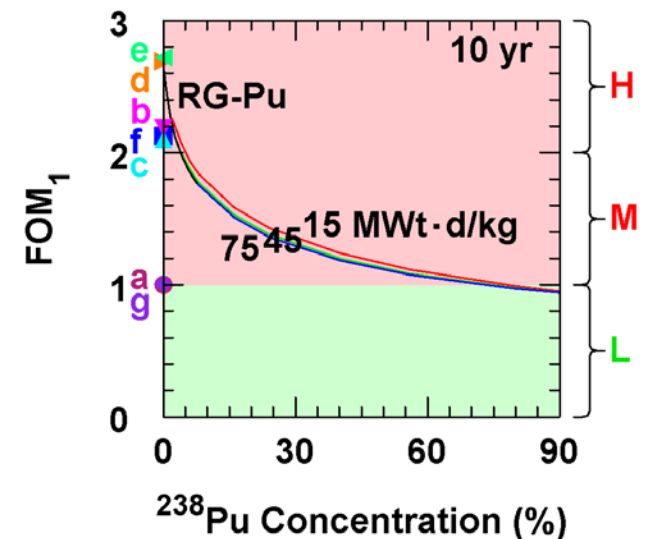
- Increasing burnup decreases the attractiveness of RG-Pu.
- Increasing cooling time increases the attractiveness of RG-Pu.
- Claims:
 - Pellaud, "The plutonium in spent fuel from light water reactors is hardly suitable for weapons utilization ..."¹
 - Kessler, "The first nuclear weapon ... did work with weapons-grade plutonium, but it would not work with reactor-grade plutonium ..."²



a – LEU (20% ²³⁵U)
 b – HEU (93% ²³⁵U)
 c – ²³⁷Np
 d – ²³³U (10 ppm ²³²U)
 e – WG-Pu (94% ²³⁹Pu)
 f – RG-Pu
 g – ²³⁸Pu/²³⁹Pu (80:20)

Denaturing (Spiking) requires large ($> 80\%$) amounts of ^{238}Pu to make the Pu unattractive.

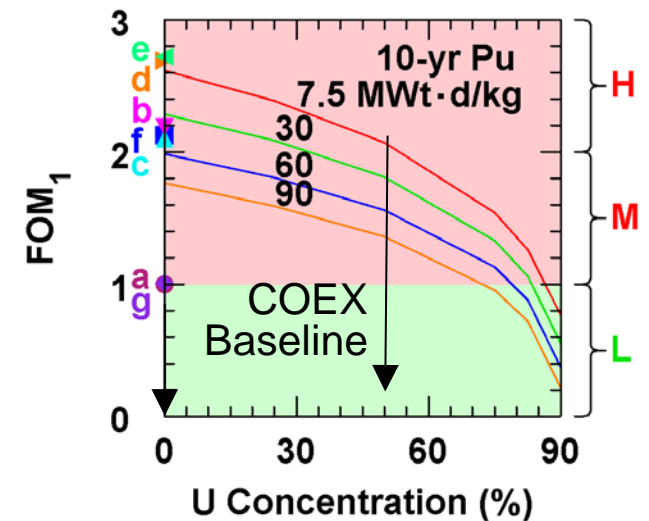
- The FOM of Pu can be reduced with:
 - higher burn-up,
 - dilution with ^{238}Pu .
- Claims:
 - Time Magazine, “The denatured [plutonium] would be harmless militarily. It would not explode.”³
 - Saito, “Since both proliferation resistant plutonium compositions [$\geq 6\%$ ^{238}Pu] and [$\geq 30\%$ ^{240}Pu] ...”⁴
 - Ronen, “We have shown that small amounts of Americium – one-tenth of one percent – is enough to obtain enough Plutonium-238 so that you cannot build a bomb.”⁵



a – LEU (20% ^{235}U)
 b – HEU (93% ^{235}U)
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Diluting reprocessed Pu metal with U metal reduces the attractiveness of resulting metal alloy.

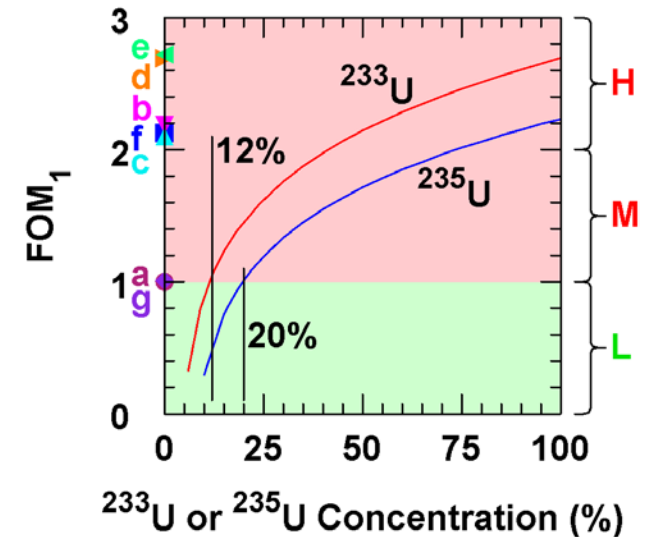
- For 10-yr, 45-MWt·d/kg Pu, $FOM_1 < 1$ requires $\geq 80\%$ U.
- DU or NU are as effective diluents as the irradiated U used herein.
- Dilution is effective only against adversaries without reprocessing capability.
- Claims:
 - AREVA initially claimed that a 50:50 mixture of plutonium and uranium produced by their COEX process was unattractive.⁶



a – LEU (20% ^{235}U)
 b – HEU (93% ^{235}U)
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 f – RG-Pu
 g – $^{238}\text{Pu}/^{239}\text{Pu}$ (80:20)

^{233}U is as attractive as weapons grade Pu (WG-Pu) and more attractive than ^{235}U .

- Note that most of the separative work has been done for ^{233}U - ^{238}U or ^{235}U - ^{238}U mixtures when they are enriched to the point of becoming attractive (i.e., $\text{FOM}_1 > 1$).



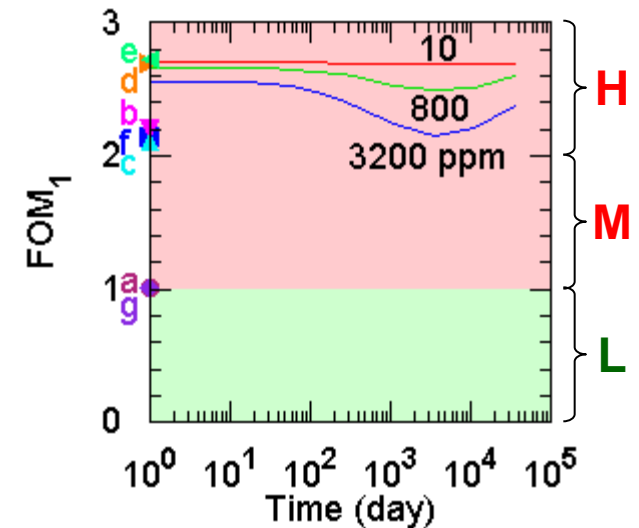
Isotope	Density (g/cm ³)	M (kg)	Mh (W)	Dose [†] (rad/h)	FOM_1
^{232}U	18.95	6.7	$4.76(10)^3$	$2.82(10)^{-1}$	1.0
^{233}U	18.95	15.3	$4.30(10)^0$	$1.46(10)^{-4}$	2.7
^{235}U	18.95	46.5	$2.79(10)^{-3}$	$1.04(10)^{-5}$	2.2
^{238}Pu	19.84	9.7	$5.51(10)^3$	$2.11(10)^{-1}$	0.9
^{239}Pu	19.84	10.0	$1.92(10)^1$	$3.95(10)^{-4}$	2.8

[†] measured at 1 m for 0.2M

- a – LEU (20% ^{235}U)
- b – HEU (93% ^{235}U)
- c – ^{237}Np
- d – ^{233}U (10 ppm ^{232}U)
- e – WG-Pu (94% ^{239}Pu)
- f – RG-Pu
- g – $^{238}\text{Pu}/^{239}\text{Pu}$ (80:20)

The attractiveness of ^{233}U is affected by its level of contamination with ^{232}U (cont.).

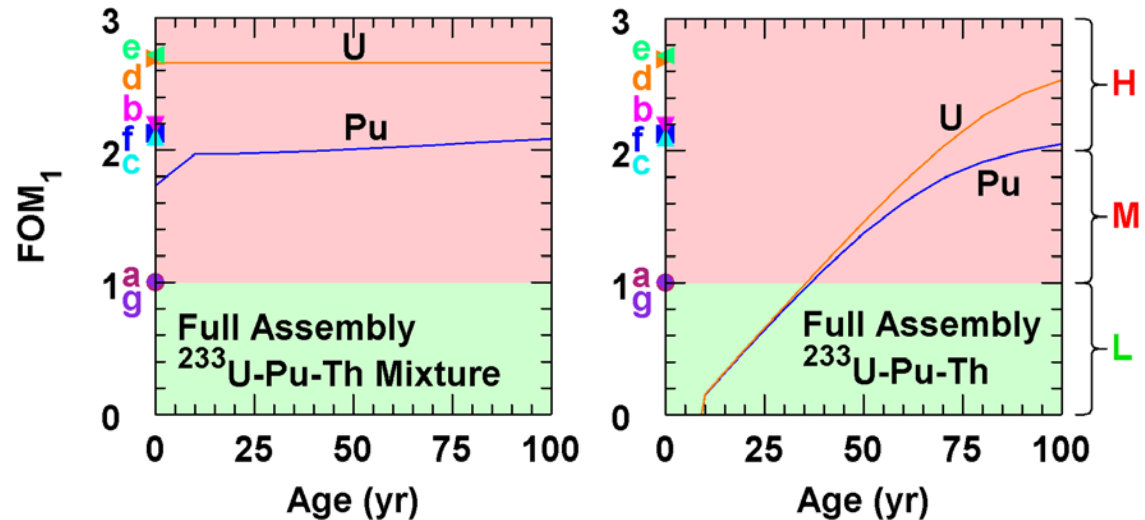
- The ^{233}U bred in reactors from thorium typically has a ^{232}U concentration of 700 to 1000 ppm.
- Even at ^{232}U concentrations of 3200 ppm, the bred uranium is highly attractive.
- Claims:
 - Sen. Hatch, “We have abundant domestic supplies of thorium, and when used in a nuclear reactor, thorium is nonproliferative ...”¹¹
 - Thorium Energy Security Act of 2010, “... nuclear power plants operating on an advanced thorium fuel cycle to generate nuclear energy – (A) would not produce weapons-usable material in spent fuel;”¹²
 - Chairman and Sec. Banerjee claimed that the contamination of ^{233}U with ^{232}U makes the bred uranium not weapons usable.”¹³



a – LEU (20% ^{235}U)
 b – HEU (93% ^{235}U)
 c – ^{237}Np
 d – ^{233}U (10 ppm ^{232}U)
 e – WG-Pu (94% ^{239}Pu)
 f – RG-Pu
 g – $^{238}\text{Pu}/^{239}\text{Pu}$ (80:20)

The attractiveness of material is dependent on the form obtained by an adversary.

a – LEU (20% ^{235}U)
 b – HEU (93% ^{235}U)
 c – ^{237}Np
 d – ^{233}U (10 ppm ^{232}U)
 e – WG-Pu (94% ^{239}Pu)
 f – RG-Pu
 g – $^{238}\text{Pu}/^{239}\text{Pu}$ (80:20)



- For adversaries with reprocessing capability, material attractiveness is given by FOM plot on left (assumes theft of reprocessed spent fuel).
- For adversaries without reprocessing capability, material attractiveness is given by FOM plot on right (assumes theft of spent fuel).
- Claims:
 - Bernstein, "...spent fuel and immobilized plutonium will remain self-protecting for about one to two hundred years."¹⁴

Questions & Answers

- Is reactor-grade plutonium attractive for use in a nuclear explosive device? **Yes, but requires processing.**
- Does co-extracting Pu with other actinides render the product unattractive? **No. However, co-extracting Am with Cm does produce a product that is unattractive.**
- At what point does dilution render Pu or a transuranic mixture unattractive?
 - Pu + U — > 80% ^{238}U concentration
 - TRU + U — > 75% ^{238}U concentration
 - TRU + Ln — > 20% of all Lanthanides in spent fuel
- Do other fuel cycles produce attractive products? **Yes, the thorium fuel cycle produces ^{239}Pu and ^{233}U . And MOX recycle produces attractive Pu.**

Generic Conclusions

- There is a safeguards and security benefit with respect to safeguards to diluting the reprocessing end products with:
 - Ln
 - U – reprocessed, natural, or depleted
- However – There is no silver bullet to solve the safeguards and security issue. None of the proposed flow-sheets examined to date justify reducing international safeguards or physical security protection levels. All reprocessing products evaluated need to be rigorously safeguarded and provided the highest levels of physical protection.

Slides for Use During Questions

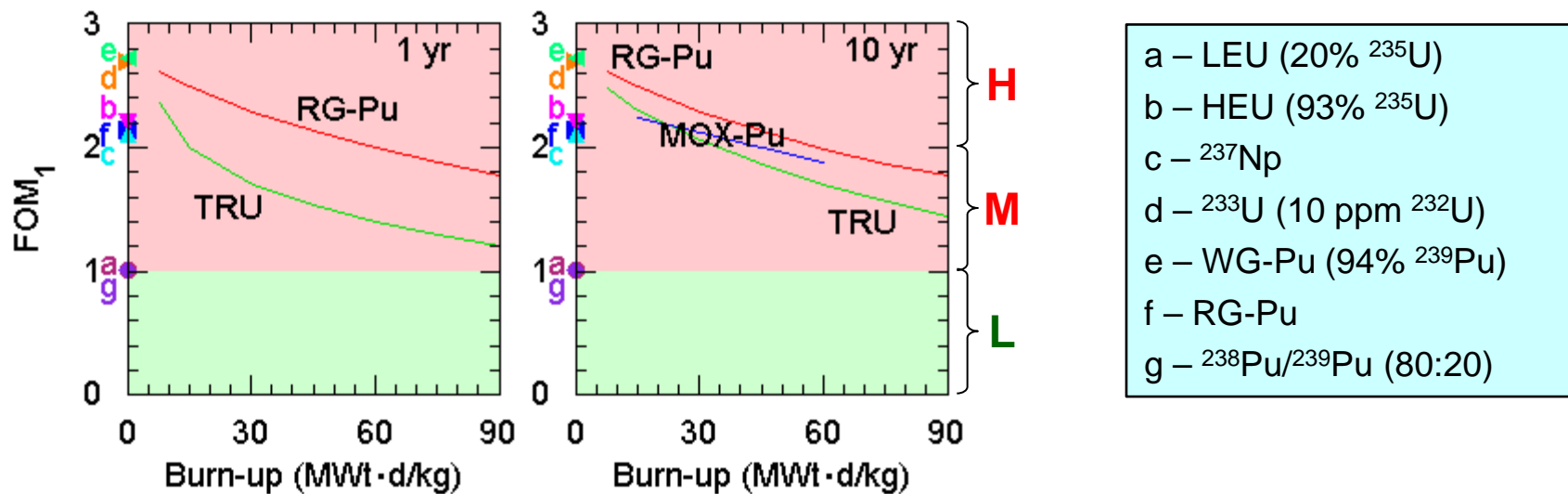
Attractiveness of Various Actinide Isotopes.

Isotope	M (kg)	Mh (W)	D [†] (rad/h)	MS (n/s)	FOM ₁	Isotope	M (kg)	Mh (W)	D [†] (rad/h)	MS (n/s)	FOM ₁
²²⁹ Th	2780.3	1.69E+04	6.18E+00	0.0	0.1	²⁴² Am	10.9	1.01E+07	1.29E+05	0.0	-6.4
²³² Pa	105.3	2.96E+08	3.64E+08	0.0	-18.8	^{242m} Am	11.7	4.94E+01	2.74E-01	1.66E+06	2.6
²³² U	6.7	4.76E+03	2.82E-01	8.74E+03	1.0	²⁴³ Am	144.8	9.31E+02	2.82E-01	4.84E+05	1.4
²³³ U	15.3	4.30E+00	1.46E-04	1.32E+01	2.7	²⁴² Cm	368.2	4.45E+07	8.55E+02	7.73E+12	-3.0
²³⁴ U	126.1	2.26E+01	3.59E-04	6.33E+02	1.8	²⁴³ Cm	11.9	2.25E+04	1.37E+02	3.18E+06	0.3
²³⁵ U	46.5	2.79E-03	1.04E-05	1.39E+01	2.2	²⁴⁴ Cm	27.1	7.66E+04	1.40E+01	2.92E+11	-0.2
²³⁶ Np	7.0	1.88E-01	1.10E-02	0.0	3.1	²⁴⁵ Cm	9.5	5.43E+01	1.47E-01	1.06E+06	2.6
²³⁷ Np	62.8	1.26E+00	4.69E-04	7.16E+00	2.1	²⁴⁶ Cm	49.4	4.93E+02	1.67E+01	4.40E+11	1.8
²³⁶ Pu	7.2	1.31E+05	6.98E+00	2.53E+08	-0.5	²⁴⁷ Cm	8.4	2.42E-02	1.62E-03	0.0	3.0
²³⁸ Pu	9.7	5.51E+03	2.11E-01	2.51E+07	0.9	²⁴⁸ Cm	42.5	5.06E+00	7.25E+01	1.85E+12	2.2
²³⁹ Pu	10.0	1.92E+01	3.95E-04	2.17E+02	2.8	²⁵⁰ Cm	24.8	3.66E+03	5.04E+03	1.64E+14	-2.0
²⁴⁰ Pu	37.3	2.64E+02	7.17E-03	3.81E+07	2.0	²⁴⁹ Bk	193.7	6.20E+04	7.49E-01	1.94E+10	-0.1
²⁴¹ Pu	13.0	4.27E+01	1.45E-03	6.50E+02	2.6	²⁴⁹ Cf	7.2	1.10E+03	5.36E+01	1.88E+07	1.6
²⁴² Pu	89.1	1.04E+01	5.45E-03	1.53E+08	1.9	²⁵⁰ Cf	6.6	2.64E+04	2.95E+03	7.37E+13	-0.7
²⁴⁴ Pu	256.2	1.29E-01	1.50E-02	4.64E+08	1.5	²⁵¹ Cf	5.6	3.16E+02	1.59E+00	0.0	2.1
²⁴¹ Am	57.3	6.55E+03	1.22E+00	6.76E+04	0.8	²⁵² Cf	5.8	1.12E+05	5.77E+05	1.37E+16	-8.2

† measured at 1 m for 0.2M

RG-Pu, irradiated MOX, and TRU are attractive.†

- Age has a significant effect on the attractiveness of TRU due to decay of ^{242}Cm and ^{244}Cm
- Age does not have a significant effect on the attractiveness of RG-Pu.

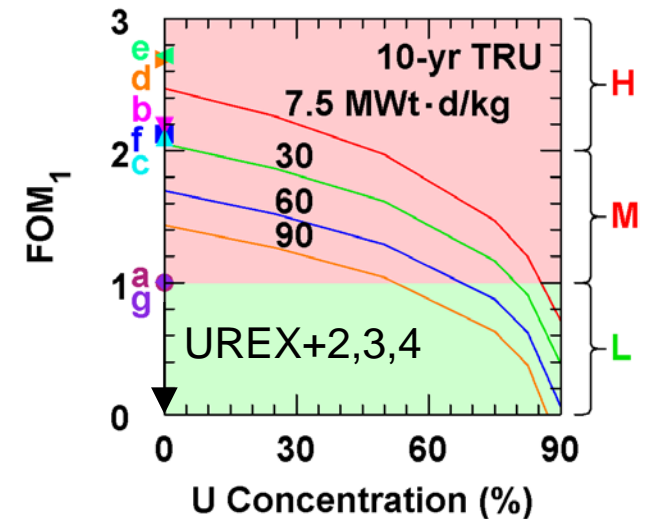


†C. G. Bathke, *et al.*, "An Assessment of the Attractiveness of Material Associated with a MOX Fuel Cycle from a Safeguards Perspective," *Proc. of INMM 50th Annual Meeting*, 2009, Tucson, AZ.

C. G. Bathke, *et al.*, "The Attractiveness of Materials in Advanced Nuclear Fuel Cycles for Various Proliferation and Theft Scenarios," *Proc. of Global 2009*, 2009, Paris, France.

Diluting reprocessed TRU metal with U metal reduces the attractiveness of resulting alloy.

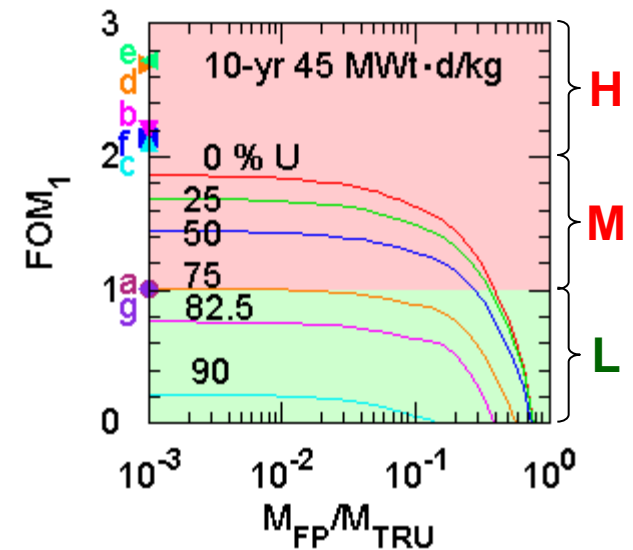
- For 10-yr, 45-MWt·d/kg TRU, $FOM_1 < 1$ requires $\geq 75\%$ U.
- DU or NU are as effective diluents as the irradiated U used herein.
- Dilution is effective only against adversaries without reprocessing capability.
- Claims:
 - Sell, "...one called UREX Plus – which, instead of separating out pure plutonium combines the plutonium with other actinides and some portion [of] uranium so that it is not attractive or usable as weapons material."⁷



a – LEU (20% ^{235}U)
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The attractiveness of pyroprocessing product is similar to TRU.

- The PYROX product can also be diluted with uranium to reduce attractiveness.
- Increasing the relative fission product mass also reduces attractiveness.
- Dilution with inert and/or radioactive material is effective only against adversaries without reprocessing capability.
- Claims:
 - Magill, “[Pyroprocessing] promises ... proliferation resistance (no separation of the TRUs)...”⁸
 - Hannum, Marsh, and Stanford, “The combination of fission products and transuranics [produced by the pyroprocess] is unsuited for weapons...”⁹

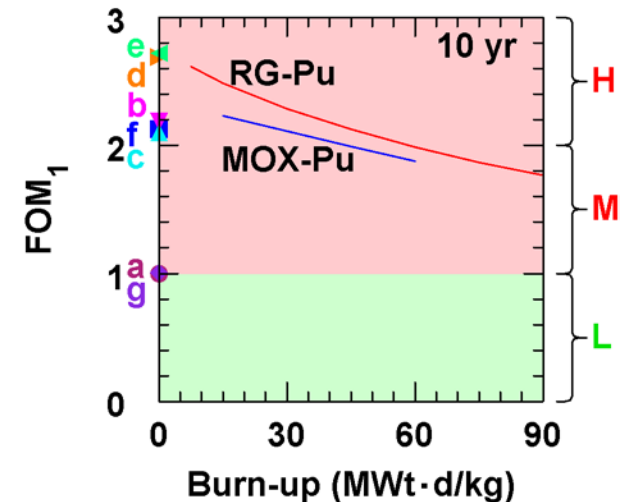


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 g – ²³⁸Pu/²³⁹Pu (80:20)

Recycling Pu as MOX consumes ~30% of the initial Pu, but only slightly reduces Pu attractiveness.

- Reprocessed Pu is an end product of:
 - PUREX
 - COEX
 - UREX+2,3, and 4.
- The FOM of Pu can be reduced with:
 - Higher burn-up,
 - Additional burn-up as MOX.
- Claims:
 - Pellaud, “The plutonium in spent fuel from light water reactors is hardly suitable for weapons utilization ...”¹⁰
 - Pellaud, “In a nutshell, MOX-recycled is not suitable for making weapons.”¹⁰

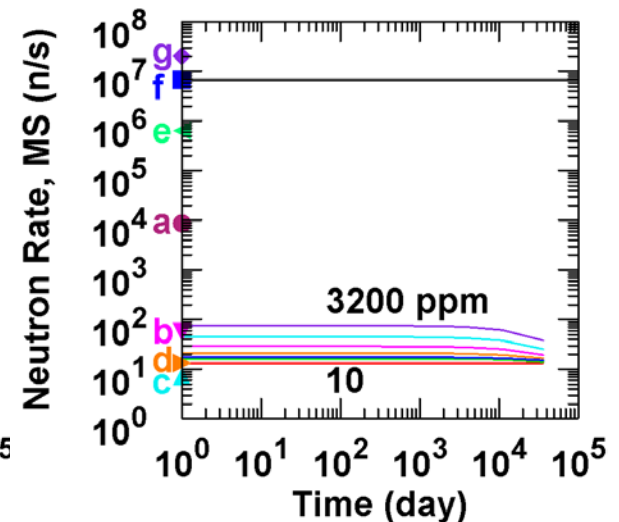
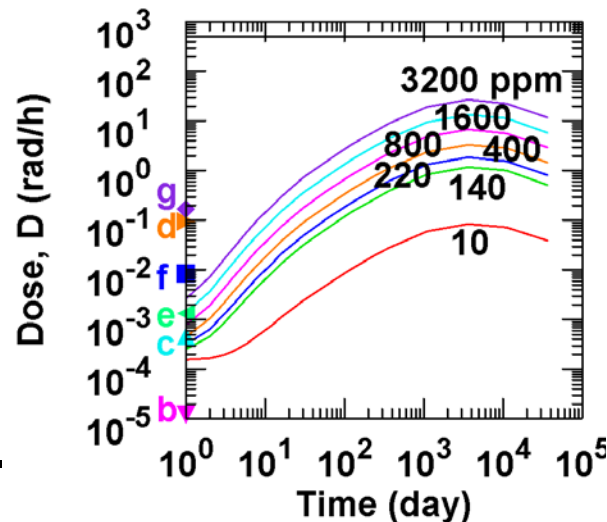
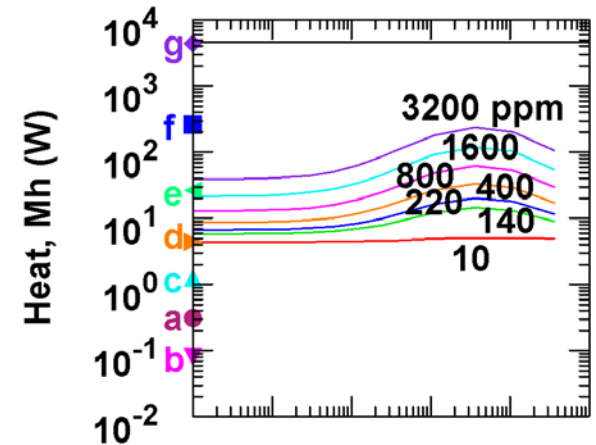
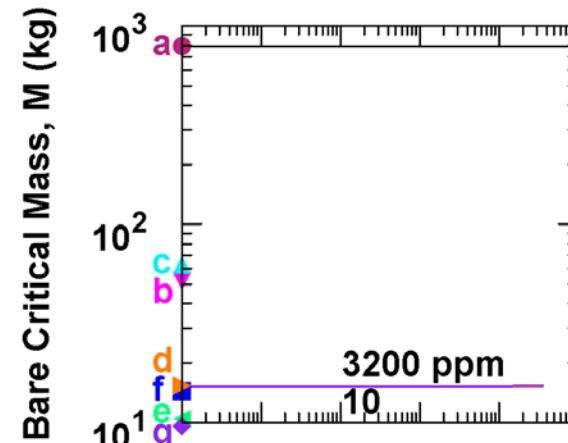
$7.5 \leq \text{UOX burn-up} \leq 90 \text{ MWt-d/kg}$
 $\text{MOX burn-up} = 60 \text{ MWt-d/kg}$



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The attractiveness of ^{233}U is affected by its level of contamination with ^{232}U .

- Typically, Th-fueled power reactors generate a ^{232}U concentration of 700 to 1000 ppm.
- ^{232}U has a 68.9 yr (25,149 day) half-life.
- Dose comes from 2.6 MeV gamma emitted by ^{208}Tl in decay chain.



Compositions of Plot Reference Points

Reference Material		Density (g/cm ³)	Composition (%)				
Letter	Descriptor						
a	LEU (20%)	18.95	²³⁵ U	²³⁸ U			
			20	80			
b	HEU (93%)	18.95	²³⁴ U	²³⁵ U	²³⁸ U		
			0.82	93.50	5.68		
c	²³⁷ Np	20.25	²³⁷ Np				
			100				
d	²³³ U (initially 10 ppm ²³² U and cooled 10 years)	18.95	²⁰⁸ Tl	²⁰⁹ Tl	²⁰⁸ Pb	²⁰⁹ Pb	²¹² Pb
			2.37(10) ⁻¹¹	4.82(10) ⁻¹⁴	6.07(10) ⁻⁵	2.01(10) ⁻¹⁰	1.40(10) ⁻⁸
			²⁰⁹ Bi	²¹² Bi	²¹³ Bi	²¹² Po	²¹³ Po
			1.83(10) ⁻⁶	1.32(10) ⁻⁹	4.72(10) ⁻¹¹	7.00(10) ⁻²⁰	7.08(10) ⁻²⁰
			²¹⁶ Po	²¹⁷ At	²²⁰ Rn	²²¹ Fr	²²⁴ Ra
			5.57(10) ⁻¹⁴	5.67(10) ⁻¹⁶	2.10(10) ⁻¹¹	5.15(10) ⁻¹²	1.22(10) ⁻⁷
			²²⁵ Ra	²²⁵ Ac	²²⁸ Th	²²⁹ Th	²³² U
			2.33(10) ⁻⁸	1.57(10) ⁻⁸	2.37(10) ⁻⁵	4.29(10) ⁻³	9.08(10) ⁻⁴
			²³³ U				
e	WG Pu	19.84	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu
			0.010	94.026	5.814	0.130	0.020
f	RG Pu (45 MWt·d/kg and cooled 10 years)	19.84	²³⁸ Pu	²³⁹ Pu	²⁴⁰ Pu	²⁴¹ Pu	²⁴² Pu
			2.56	53.16	27.73	9.52	7.02
			²⁴⁴ Pu				
g	²³⁸ Pu/ ²³⁹ Pu (80:20)	19.84	²³⁸ Pu	²³⁹ Pu			
			80	20			

Details of FOM Calculations for Plot Reference Points

Material		M (kg)	α_{∞} (gen/shake)	Mh (W)	Dose [†] (rad/h)	MS (n/s)	FOM ₁
Letter	Symbol						
a	LEU (20%)	771.8	0.16	$1.45(10)^{-2}$	$1.21(10)^{-5}$	$8.44(10)^3$	1.01
b	HEU (93%)	51.8	1.13	$7.92(10)^{-2}$	$1.21(10)^{-5}$	$5.66(10)^1$	2.18
c	^{237}Np	62.8	1.55	$1.26(10)^0$	$4.69(10)^{-4}$	$7.16(10)^0$	2.10
d	^{233}U (10 ppm)	15.3	2.45	$4.59(10)^0$	$8.30(10)^{-2}$	$1.33(10)^1$	2.69
e	WG-Pu	10.5	3.09	$2.35(10)^1$	$5.78(10)^{-3}$	$6.70(10)^5$	2.73
f	RG-Pu	14.4	2.69	$2.57(10)^2$	$8.42(10)^{-3}$	$6.75(10)^6$	2.13
g	^{238}Pu (80%)	9.7	3.37	$4.39(10)^3$	$1.67(10)^{-1}$	$2.00(10)^7$	1.01

† measured at 1 m for 0.2M

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